## Supporting Information

## Encapsulation of Dyes in Metal-Organic Frameworks and the Tunable Nonlinear Optical Properties

Tao Song, Jiancan Yu, Yuanjing Cui,\* Yu Yang and Guodong Qian\*

State Key Laboratory of Silicon Materials, Cyrus Tang Center for Sensor Materials and Applications, School of Materials Science and Engineering, Zhejiang University, Hangzhou, 310027 (China)

E-mail: cuiyj@zju.edu.cn (Y. Cui); gdqian@zju.edu.cn



Figure S1. <sup>1</sup>H NMR spectra (500 MHz) of the dye (a) MBPDC and (b) DMBPDC



Figure S2. TGA curves of bio-MOF-Me and bio-MOF-2Me



$$\begin{split} S_{BET} = 1/(Slope+Intercept)/22414 \times 6.023 \times 10^{23} \times 0.162 \times 10^{-18} = 1011.4 \text{ m}^2/\text{g} \\ \textbf{Figure S3.} \text{ The BET surface area of bio-MOF-1 obtained from N}_2 \text{ adsorption} \\ \text{ isotherm at 77 K} \end{split}$$



 $S_{BET}=1/(Slope+Intercept)/22414 \times 6.023 \times 10^{23} \times 0.162 \times 10^{-18} = 868.7 \text{ m}^2/\text{g}$ Figure S4. The BET surface area of **bio-MOF-Me** obtained from N<sub>2</sub> adsorption isotherm at 77 K



 $S_{BET} = 1/(Slope+Intercept)/22414 \times 6.023 \times 10^{23} \times 0.162 \times 10^{-18} = 745.2 \text{ m}^2/\text{g}$ 



isotherm at 77 K



**Figure S6.** Polarized fluorescence spectra of (a) bio-MOF-1⊃DM-1 and (b) bio-MOF-1⊃DP-1



**Figure S7.** The polarized absorption spectra of (a) bio-MOF-1⊃DM-1 and (b) bio-MOF-1⊃DP-1, and the corresponding dichroic ratio (black dots).



Figure S8. Polarized absorption spectra of (a) bio-MOF-1⊃DM-1, (b) bio-MOF-1⊃DM-4, (c) bio-MOF-1⊃DM-7 and (d) bio-MOF-1⊃DM-7. The legend means the angle between the polarizer and the long axis of crystal



Figure S9. Polarized absorption spectra of (a) bio-MOF-Me⊃DM-1, (b) bio-MOF-Me⊃DM-4, (c) bio-MOF-Me⊃DM-7 and (d) bio-MOF-Me⊃DM-7.



Figure S10. Polarized absorption spectra of (a) bio-MOF-2Me⊃DM-1, (b) bio-MOF-2Me⊃DM-4, (c) bio-MOF-2Me⊃DM-7 and (d) bio-MOF-2Me⊃DM-7.



Figure S11. Polarized absorption spectra of (a) bio-MOF-1⊃DP-1, (b) bio-MOF-1⊃DP-4, (c) bio-MOF-1⊃DP-7 and (d) bio-MOF-1⊃DP-7.



Figure S12. Polarized absorption spectra of (a) bio-MOF-Me⊃DP-1, (b) bio-MOF-Me⊃DP-4, (c) bio-MOF-Me⊃DP-7 and (d) bio-MOF-Me⊃DP-7.



Figure S13. Effect of reaction time (the right one describes the pseudo-second-order kinetic) on DM-1 adsorption onto (a) bio-MOF-1, (b) bio-MOF-Me and (c) bio-MOF-2Me



Figure S14. SHG spectra of DM-1 crystals